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# FOG DROP SIZE DISTRIBUTIONS - MEASUREMENT METHODS AND EVALUATION

By

Gayle S. Rinehart

JUL 25 1969

**ATMOSPHERIC SCIENCES LABORATORY**  
WHITE SANDS MISSILE RANGE, NEW MEXICO

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## ABSTRACT

In connection with evaluating fog modification efforts, several published methods of measuring the size distribution of fog droplets were reviewed. An evaluation cast doubt on the validity of the gelatin, Formvar, and polyvinyl alcohol, and oil collection media for recording droplets less than  $4\mu$  diameter. Efficiency of several collection methods appeared to be poor for less than  $4\mu$  diameter droplets when lack of drop size distribution correlation with visibility measurement was considered.

During a fog dispersal test, pyrotechnically produced hygroscopic reagents were observed to cause an increase in the number of concentrations of the small sized droplets of a natural fog.

During nonfog conditions, several types of pyrotechnic flares containing different hygroscopic reagents were tested for usefulness as fog modification agents. Measurement of drop and particle size distributions downwind from the ignition point of these pyrotechnics revealed that the effect of small droplet sizes in decreasing visibility in air of high humidity would negate the improvement made by removal of large size drops.

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## INTRODUCTION

To evaluate methods of increasing visibility in or dispersing warm (above 0°C) fog, knowledge of the change in physical properties of the fog is imperative. One of the most meaningful physical properties is the drop size distribution, because the greatest visibility impairment in fog results from light scattering by water droplets. Since scattering cross-section coefficients are drop-size dependent, the change in drop size distribution after a modification experiment is of paramount interest.

To evaluate the possible methods of determining drop size distribution for purposes of evaluating fog modification techniques, published methods were surveyed. This survey revealed that drop size determinations were somewhat method-dependent. To estimate the validity of various published distributions and their dependence on measurement methods, a laboratory study of four types of collecting media and three collection methods was undertaken.

In light of knowledge gained from these experimental results, published values of the number and sizes of natural fog droplets were reevaluated. Basis for acceptance and rejection of the number of droplets in the various size increments, as valid measurements, is discussed.

Knowing the limitations of techniques for capturing and measuring fog drops, one impactor method employing oil as a recording medium and the natural settling method employing gelatin and Formvar as media were selected as the best combination of the capture methods and recording media examined. The suitability of the above capture method and media combinations was tested in May and June of 1968 on a radiation fog at Ft. Rucker, Alabama, and an advection fog at Arcata, California, in conjunction with fog modification experiments.

To investigate the growth and fall-out of the drops resulting from the condensation of water on nuclei created by pyrotechnic flares, a follow-up experiment was carried out. Four types of pyrotechnic flares were burned in nonfog conditions and size distributions measured downwind at three distances from the source of the nuclei. From this study the effectiveness of these pyrotechnics in dispersing fog was extrapolated. This report summarizes the results of these studies in three parts.

In Part I, results of laboratory studies on published droplet capture methods and recording media for determining fog drop size distributions are presented and discussed. The information obtained from these studies was incorporated in the evaluation process for establishing the validity

of published fog drop size distributions as outlined and discussed in Part II.

In Part III, results of fog drop size distribution and visibility measurements of a radiation fog are discussed. Measurements made before and during a weather modification experiment employing pyrotechnically produced hygroscopic nuclei are presented and evaluated. During nonfog conditions, four types of pyrotechnics which produced hygroscopic reagents were compared for suitability for weather modification applications.

## PART I

### DROPLET CAPTURE METHODS AND RECORDING MEDIA

Although an annotated bibliography of methods of collecting and determining fog and rain drop distributions exists (1), few of these methods are applicable to capturing or sizing fog droplets below  $20\mu$  diameter. A survey of these and more recent approaches to determining fog drop size distributions was carried out in this laboratory as an insight to judging the value of various approaches.

#### A. Review and Discussion

Four general types of media for directly capturing or replicating fog droplets smaller than  $20\mu$  diameter are as follows:

- (1) Capturing and measuring replicas by means of plastic (Formvar).
- (2) Capturing and measuring impressions made in a layer of small particles (magnesium oxide, soot) or an amorphous solid (i.e., gelatin, polyvinyl alcohol, dye stain).
- (3) Capturing in fluid (oil, silicon fluid, petroleum jelly).
- (4) Capturing on spider webs or wire.

#### Replication in Formvar

A 5% solution of Formvar in ethylene dichloride or chloroform for use as a fog droplet capture medium is employed as follows. A layer of Formvar/solvent solution is spread on a flat collecting surface. The layer is usually allowed to dry and is then remoistened with solvent just before use. As droplets hit and penetrate the layer, the solvent evaporates causing the Formvar plastic to harden around the captured droplets. Eventually the water from the droplets evaporates through the Formvar leaving behind a permanent replica of the droplets.

The size of the droplets has been estimated by multiplying the replica by a factor of 0.5 for large drops (2). Small drops of diameter thinner than the Formvar layer do not spread but evaporate eluding a standard correction factor (3). The number of droplets captured has been related to the number existing in an aspirated volume of air by applying a factor to allow for the collection efficiency of various



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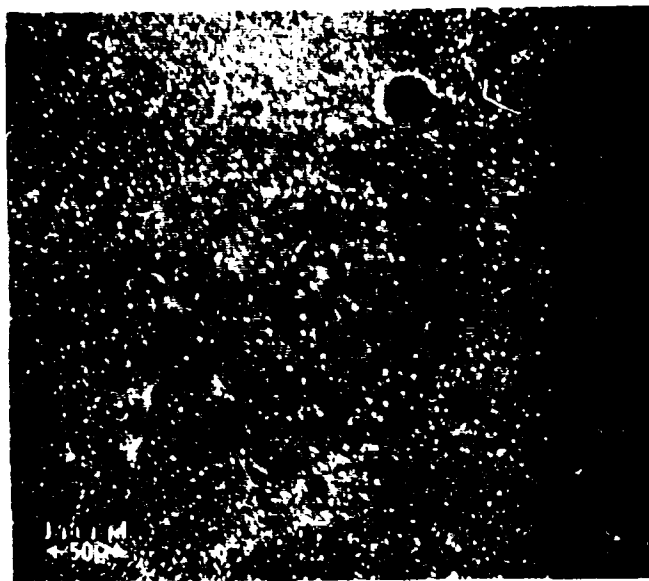


FIGURE 1. FORMVAR REPLICAS OF NATURAL FOG FROM ARCATA, CALIFORNIA, SHOWING BACKGROUND BLUSHING.

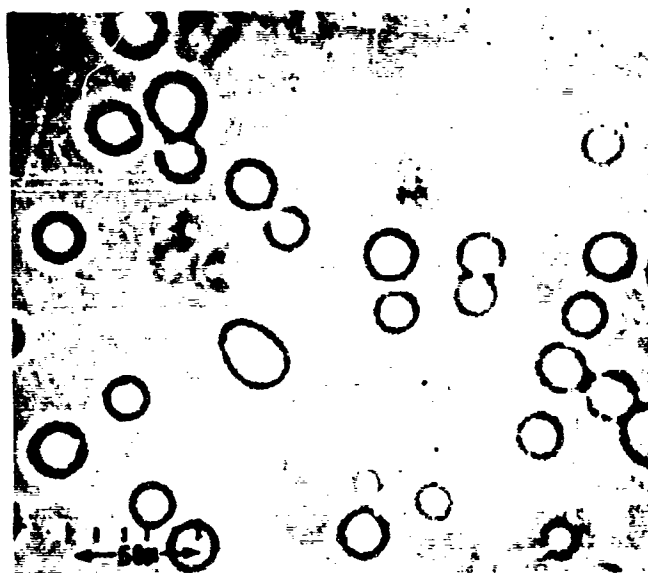


FIGURE 2. IMPACTED SYNTHETIC FOG DROPS ON FRESH KNOX GELATIN.

sized droplets (4); however, later investigations have shown the capture efficiency for small droplets to be much higher (3). Although air is usually aspirated over the collection surface or the collector is moved by aircraft through air, gravity settling has also been employed (5). The volume of air through which collected droplets have fallen during the slide exposure time can be calculated by considering the terminal velocity of the collected droplets and the sampling area.

Because of blushing, the practical lower limit of distinguishing fog droplets by employing the Formvar method in most cases is near  $2\mu$  diameter (6). Blushing is caused by the presence of tiny droplets of about 1 or  $2\mu$  diameter, which result from local cooling due to the evaporation of the solvent. The cooling hardens the Formvar, possibly trapping bubbles of solvent vapor as they rise through the medium. Examples of fog droplets (dark rings) and blushing (light dots) are shown in Figure 1.

#### Impressions in Layer of Small Particles or Amorphous Film

The first use of the small particle film method is attributed to May (7) who coated microscope slides with a magnesium oxide film by waving the slide over burning magnesium ribbon. Fog droplets cause craters in the film upon impaction; these can be related to droplet size by multiplying by a spread factor. It was found that droplets were reliably detectable only down to  $10\mu$  according to May (8) and  $25\mu$  diameter according to Rief and Mitchell in a more recent study (9) because of the apparent hardening of the magnesium oxide layer.

An amorphous film of methylene blue on microscope slides (9) and a water blue film on nitrocellulose (10) have been employed. Correction factors for sizes of drops falling under gravity on slide and film were 0.42 (9) and from 0.5 to 0.33 times the observed size depending on the drop size (10).

Gelatin layers have been employed to size droplets in a number of cases (11, 12, 13, 14). The 5% gelatin solution is warmed, spread on slides, and allowed to dry. Impinging droplets hit the gelatin surface and cause craters which can be viewed clearly under phase contrast microscopy (Figure 2). Natural settling of droplets on gelatin coated slides has been employed by Hosler (14) who approximated the spread factor correction under natural fall as 0.5 to 1.0 times observed. Mechanical impactors with gelatin slides have been used by Liddell and Wooten (11), May (12), and Meszaros (13). Velocities in the Meszaros impactor require a size correction of 0.7 times observed; while the Casella impactor (designed by May) requires 0.55 to 0.65 (7).

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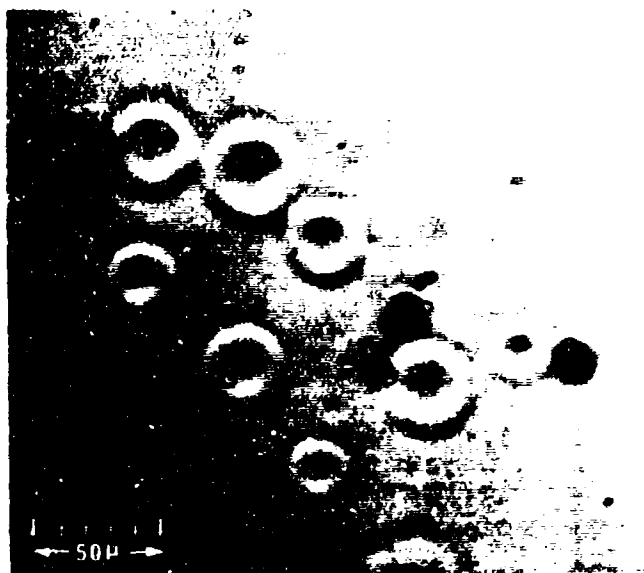


FIGURE 3. WATER DROP REPLICAS (WHITE CRATERS) AND CHLORIDE DROP REPLICAS (DARK CIRCLES) ON SENSITIZED POLYVINYL ALCOHOL.

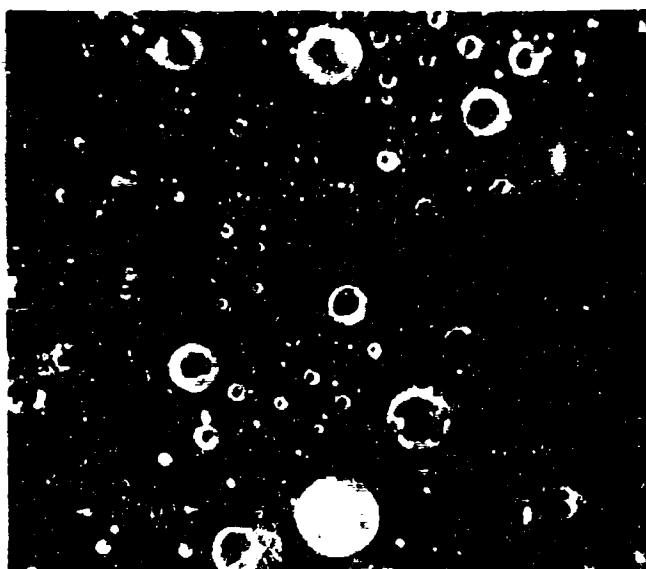


FIGURE 4. SYNTHETIC FOG DROPLETS CAPTURED IN SILICON OIL.

Polyvinyl alcohol (PVA) is used in a manner similar to gelatin. The PVA is dissolved in a water solution of silver nitrate and hydrogen peroxide (15). The solution is applied to clear Kronar film (Eastman Kodak Co.) by dipping and drying. The film thus prepared is sensitive to approximately 0.02% chloride droplets (dark craters in Figure 3) with droplet detection down to  $1\mu$  diameter. The PVA film is superior to gelatin in crater formation and humidity resistance and has a spread factor of 0.56 (16). An explanation of the mechanism of PVA-film darkening of chloride drops is given by Koenig (17).

Collargol has been employed in a single stage impactor by Dessens (18) and by Godard (19). The collargol films must be prepared 5 to 10 minutes before use, and impressions of good quality can be obtained without the use of phase contrast microscopy when freshly prepared collargol slides are used. The impression diameter when multiplied by 0.34 yields the actual drop diameter and the detection limit is  $0.5\mu$  diameter.

#### Capturing in Fluid

Fluid has been used as a collector medium by Diem (20), May (7), Okita (21, 22), Keily (23), and Kumai and Francis (24). Diem used mineral oil, Okita employed cedar oil, and Kumai and Francis used silicon fluid. Others used oil of an unspecified type. The capturing efficiency of a variety of oils is not good when employing gravity settling (9) because of low droplet penetration and lengthy sampling periods. The droplets evaporate into the oil medium necessitating quick sample study. The size detection limit in oil is specified by Keily (23) as  $2\mu$  diameter; however, the author believes that the limit is equal to the limits of the optical microscope (somewhere between  $0.5\mu$  and  $1.0\mu$  diameter).

An advantage of the use of fluid lacking in replication methods is the clarity of the spheres (Figure 4) and the distinguishability between small droplets and particles which is not found in gelatin. No size correction factor is required in fluid since there is no droplet distortion. The disadvantages in using fluid capture are the lack of a permanent sample, poor droplet penetration into fluid, and the possible coalescence of the droplets.

#### Collection on Spider Webs

Dessens (25) and Arnulf, et al. (26) employed spider webs to capture small droplets in fogs, photographing them shortly after collection. Arnulf estimated that numerous particles smaller than  $4\mu$  diameter must have escaped capture by the web filaments, and this conclusion was borne out by data of Eldridge (27).

Knowledge of wind speed past the web is essential for calculating the volume of air sampled. The webs may be mounted and rotated at a known speed, thus eliminating the uncertainty associated with natural wind variability. Droplets collected on the webs must be photographed soon after collecting, preferably in an environment in which relative humidity is near that of the air sampled.

#### Collection Efficiency of Natural Settling, Impactor, and Web Methods

The collecting efficiency for the Casella cascade impactor has been documented (7), and correlations between number and size of drops and visibility have been good (28). The vacuum impactor efficiency (24) as determined by experiment indicated higher values than theoretically calculated for nearly all sizes. Small collector areas coated with oil have been found to have higher collecting efficiencies when samples are impacted than those theoretically calculated (23, 24, 29). The collection efficiency for various sized droplets when aspirated over and caught on film using Formvar replication has not been published in the open literature except when used in the MacCready device. MacCready's values (2) are in good agreement with those of Dessens (2) using the spider web technique down to  $8\mu$  diameter.

The distance between the web filaments does not permit capture of all droplets below  $15\mu$  diameter (19) and for this reason numbers of drops in sizes below this become increasingly more unreliable when employing the web technique.

Gravity collection is not efficient for small droplets because they do not fall under the force of gravity alone. Slight turbulence or wind overcomes their downward velocity component and prevents their settling. They do not penetrate oil well and evaporate from the surface, and they do not leave impressions in gelatin or PVA.

#### **B. Experimental Procedure**

Four collection media (PVA, Formvar, gelatin, and oil) were investigated employing three collecting methods (gravity settling, Casella 4-stage impactor (7), and two-stage vacuum impactor (30)). Other media were discarded because of difficult or lengthy preparation procedures, lack of sensitivity to droplets smaller than  $2\mu$  in radius, poor quality of replication, and nonpermanence of sample.

Two types of fog generating equipment were employed: an aspiration type medicinal nebulizer and an ultrasonic droplet generator (Ultramist III, Macrosonics Corp., Rahway, N. J.).

### Formvar

A standard microscope slide was coated with 5% Formvar/ethylene dichloride solution by placing a drop of the solution on the slide and spreading this coating by means of another slide. The slide was found to dry within 10 to 90 seconds, depending on the thickness of the solution. A thinly coated slide was exposed to a stream of drops of 7 to 10 $\mu$  diameter (average) generated by the ultrasonic generator for one minute while being observed under the microscope. Photographs of this slide during and after the collection period are shown in Figures 5 and 6. Figure 5A shows the slide after 10 seconds exposure and Figure 5B after 20 seconds exposure. Figures 6A and 6B show the droplets evaporating and in the final state.

The capturing of fog droplets by gravity settling onto a 5% Formvar solution did not appear to be reliable. Small drops falling on the slide early evaporated, while some larger ones (over 10 $\mu$  diameter) spread and then decreased in size. This may explain the absence of large droplets when Formvar was used in the data of Admirat and Soulage (6). Small drops (less than 5 $\mu$  radius) appeared to be unchanged upon impaction, while larger drops (over 20 $\mu$ ) spread. Since the hardening Formvar is of variable thickness, it would be expected that spread factors would also vary. After further study this was found to be true (3) even in softened Formvar. Because of the rapid drying time and the evaporation of droplets, Formvar was deemed unsuitable as a collection medium for the cascade or vacuum impactor.

### Gelatin, Polyvinyl Alcohol (PVA)

Studies on aged gelatin in field conditions showed its inability to retain impressions made by droplets (Figure 7A and 7B). Rejuvenation of old gelatin is possible by rinsing with distilled water (12). Certain gelatins seem to lose their impressionable nature faster than others, and the author has found filtered Knox unflavored gelatin (Figure 2) to be quite suitable for retaining images.

One packet of Knox unflavored gelatin is added slowly to 500 ml distilled water. The solution is warmed in a water bath at approximately 85°C with occasional stirring until all the gelatin is dissolved. The warm solution is filtered through an 0.8 $\mu$  pore size membrane filter under suction and can be reheated when required. An eyedropper is used to withdraw the gelatin solution to avoid picking up bubbles on the gelatin surface.

Natural settling of fog droplets on gelatin films did not make permanent impressions in all cases (Figure 8). This did not appear to be as

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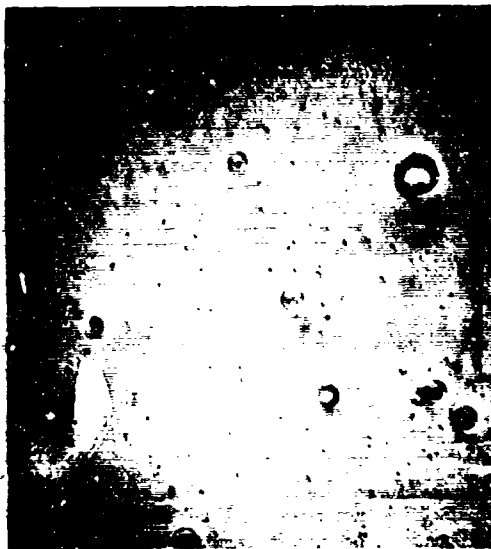


FIGURE 5. SYNTHETIC FOG DROPLETS FALLING ON THIN FORMVAR SOLUTION.

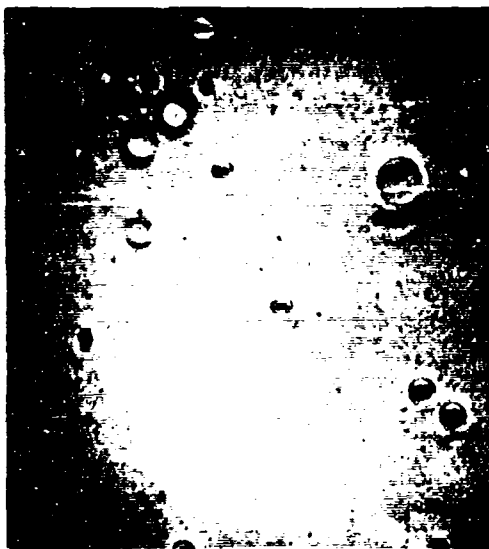
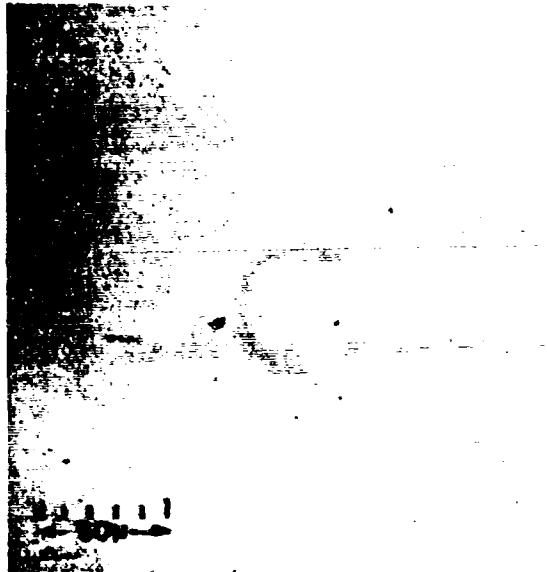


FIGURE 6. SYNTHETIC FOG DROPLET REPLICAS AFTER FORMVAR SOLUTION HAS DRIED.

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A. AT FT. RUCKER



B. AT ARCATA

FIGURE 7. POOR REPLICATION OF FOG DROPLETS ON GELATIN.



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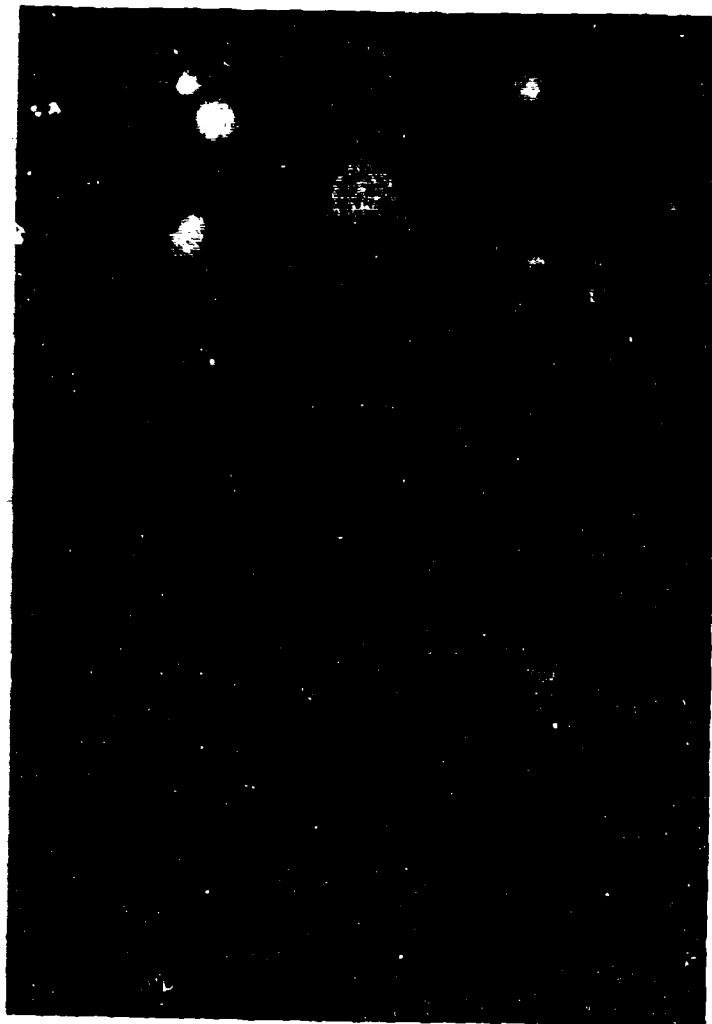


FIGURE 8. FAINT FOG DROPLET IMPRESSIONS ON GELATIN AFTER NATURAL SETTLING.

critical for larger drops or impacted samples on gelatin-coated slides; however, several faint craters are visible on an impacted sample (Figure 2), and it can be assumed that some drops were not recorded as in the case of natural settling.

PVA was found to be superior to gelatin in recording impressions of droplets under natural settling; however, not all droplets were permanently recorded (Figure 9). Some remaining yet faint impressions are shown in this figure. PVA employed in vacuum and cascade impactors appeared to record impressions better than when gravity settling was used, although faint impressions indicate that some were not recorded.

#### Silicon and Petroleum Oil

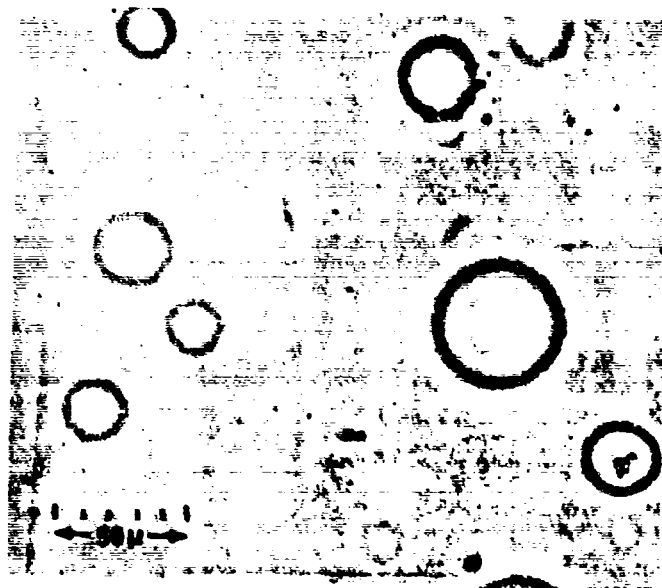
Use of petroleum oil and silicon oil for gravity settling yielded poor results owing to the evaporation of droplets on top of the oil. Even droplets suspended in oil evaporated quickly into the medium. Water-saturated petroleum oil gave better results; however, droplets still evaporated more slowly in the silicon fluid than in the saturated oil. Therefore, silicon fluid was employed in the three collection methods.

Thick silicon oil (1000 centipoise) is easier to work with because it is more viscous and can be held vertically when on a slide without running off. The sample once captured remains in a smaller area for easier microscope counting. Small droplets, however, do not penetrate thick silicon oil easily. If the velocity of the droplets to be collected is increased to allow small drops easier penetration, larger droplets in the same volume of air go completely through the oil layer and flatten against the slide.

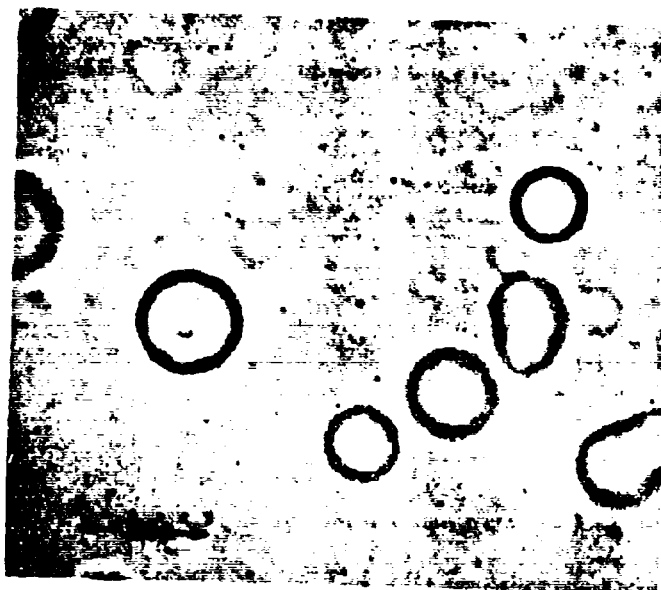
One method of relieving this problem is to place a thin layer of thick oil next to the slide and cover it with a lighter oil. This practice allows large droplets to penetrate the second layer partially and small droplets to penetrate the first layer more easily.

Since viscosity varies with temperature and the number and size of droplets in a given volume of air also vary, no standard viscosity fluids are recommended. In general, at 25°C Dow Corning No. 200 silicon fluid (1000 centipoise) at average air speeds of six m/second gives fair collection for all sizes (Figure 3). When the Casella and vacuum impactors were used with silicon fluid some losses occurred due to evaporation before slides could be removed from the instrument. An oiled cover glass was placed on the sample preventing the evaporation and flattening of droplets on the top of the oil film of the collector. Most droplets on slides preserved in this manner lasted for hours; however, small drops (1 to 2  $\mu$  diameter) evaporated after about 15 minutes.

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A. NATURAL SETTLING



B. IMPACTION

FIGURE 9. FAINT FOG DROPLET IMPRESSIONS ON PVA AFTER NATURAL SETTLING AND IMPACTION.

A short sampling time was essential when sampling with an oil medium in the Casella impactor because the droplets coalesced with others lying on the top of the oil layer, and evaporation losses were large.

The vacuum impactor had the same drawback as the Casella impactor mentioned above, but sampling was very fast and only one slide had to be removed from it, thus reducing evaporation before protecting with an oiled cover glass.

### C. Conclusions

Formvar, PVA, gelatin, collargol, fluid, and dye spread on various flat surfaces can be employed to document fog droplets. A factor must be applied for collection efficiency and spreading, except when employing oil or fluid with which no spread factor is required. Except for Formvar when blushing is present, the impression media allow measurement of droplets down to approximately  $0.5\mu$  diameter. For oil, the somewhat higher limit of measurement of  $2\mu$  diameter (23) is possible.

Short sampling periods for gelatin, PVA, and Formvar are necessary because long exposure to high relative humidity causes poor definition of craters or blushing. Short sampling periods per surface exposure are required for oil to reduce coalescence, and photographs of oil-captured drops must be made soon to record small droplets. Formvar cannot be used reliably due to evaporation of captured fog droplets and variability of spread factors during drying.

## PART II

### A COMPARISON OF PUBLISHED VALUES OF FOG DROP PARAMETERS

In light of knowledge gained in Part I of this report, a reevaluation of published values of fog droplet parameters was deemed necessary. The verification of the drop size distribution dependence on collection methods as determined in Part I is borne out by results of the Puy d'Dome Comparison Conference, 1965 (6); yet few attempts (28 and 31) have been made to elucidate the reason for noncorrelated results.

Part II of this report presents a review of published fog drop size distributions and discusses the attributes and disadvantages of each method used for the purpose of correlating fog drop parameters.

#### A. Comparison of Data

Table I gives some published fog droplet parameters. Only those data obtained with methods and media allowing droplet size collection and measurement to below  $5\mu$  radius are included in the table. Entries in the table are grouped according to the type of collecting method or media employed. The paragraphs that follow discuss the relative merits of these groups and the individual entries therein.

#### Oil

Kumai (24) found the fog drop distribution mode to approach  $2\mu$  radius, after good efficiency of a syringe-operated vacuum impactor had been shown. Diem's (20) mode for stratus was  $2\mu$  radius when employing an aircraft carried droplet pistol. Other clouds had modes of  $5\mu$  and  $10\mu$ . Minervin (29) found the mode to be near  $3.5\mu$  radius when sampling from an aircraft on an oil-coated slide, estimating that smaller sizes must have been missed. Keily (23) showed an increasing number of droplets toward lower limits of detection ( $2\mu$  diameter) when oil was employed utilizing a method having good collection efficiency. Kojima (33) noted that 3 out of 4 types of fog examined had modes below  $10\mu$  when captured in cedar oil and corrected for impaction efficiencies. In the remaining case, fog blown in from over the sea had a mode of 5 to  $10\mu$  radius.

Houghton (35) employed natural settling to capture drops listing the mode of distributions between 3 and  $8\mu$ . This author believes he probably missed many small droplets because of the inefficient gravity settling.

TABLE I. COMPARISON OF DROPLET PARAMETERS AND COLLECTION METHODS

TYPE MEDIUM	DETECTION LIMIT RADIUS ( $\mu$ )	MEDIUM: OIL (CEDAR, SILICON, PETROLEUM)				COLLECTOR - REMARKS
		MODE RADIUS ( $\mu$ )	NO./CM <sup>3</sup>	VISIBIL- ITY (m)	REF	
P	0.5	3	ng	ng	20	Aircraft droplet pistol
S	1	5-6	2.5	ng	24	Syringe impactor
S	1	2	2.5	ng	24	Syringe impactor (Sum of dist.)
ng	1	3.5	ng	ng	29	Aircraft impacted
-	1	1.5, 14	ng	ng	23	Aircraft impacted
C	1	0-5	10-	ng	32	Syringe impactor
		5-10	175		32, 33	
C	1	0-10	0.7-	ng	34	Syringe impactor
		5-10	22			
P	1	10-70				
		3-8	7-24	45.6- 744	35	Natural settling
MEDIA: GELATIN, STAIN, FORMVAR, PVA						
G	0.5	1	ng	50	11	Casella impactor
G	0.5	3-10	1-50	70-	13	Own impactor
				150		
G	0.5	Median	1-1300	75-	28	Casella impactor
		<1-2.4		400		
S	0.5	8-13.5	1-8	ng	36	Natural settling
F	2	10	ng	ng	2	Own impactor
P	1	5-60	1-70	.70-	37	Electrostatic impactor
				270		
MEDIUM OR PRINCIPLE: WEB, TRANSMISSION						
W	2	10	ng	ng	2	Web. No values below 4 $\mu$ radius
W	2	<2	250*	71	26	Web. No values below 2 $\mu$ radius
T	-	0.35	800-	ng	31	Calc. from transmission of Arnulf
			10500			
T	-	0.5-	38000	ng	27	Calc. from IR transmission
		2.5	64000			
W	2	<2	100	-	25	Web. Correlated with transmission measurements

\* According to estimated data (31).  
ng = not given

Except for sea fog, when impaction techniques of known efficiencies were used, the fog or cloud drop distribution mode approached  $2\mu$  or less. Kojima (34) found that advected sea fogs had higher modal values.

#### Gelatin, Stain, Formvar, Polyvinyl Alcohol (PVA)

Most of the users of gelatin and Formvar employed impaction methods. May (28) and Liddell and Wooten (11) found the median or mode to be from  $2.4\mu$  to less than  $1\mu$  radius. Homma (36), MacCready (2), Meszaros (13), and Evans (37) could distinguish  $1\mu$  droplets yet found modes of near  $10\mu$  radius in a number of distributions. The absence of the mode at small radii in Homma's study could be because he did not impact samples and, therefore, missed small droplets. MacCready employed Formvar but because of blushing could not see below  $2\mu$  radius; still, however, the number in the less than  $5\mu$  radius region increased toward lower limits. Meszaros (13) impacted samples on gelatin but was unable to show good correlation between visibility and observed drop size distributions, which had modes from 3 to  $10\mu$  radius. Calculated and actual visibilities differed by a factor of 10 to 20 in most cases. Conversely, May (28) found the median radius below  $2\mu$  in most cases and was usually able to correlate drop size distributions and visibility to within a factor of 2.

Meszaros (13) suggested that the lack of observation of small droplets and the presence of low droplet concentrations (1 to 10 drops  $\text{cm}^{-3}$ ) could be due to local conditions. She used the Casella (May) impactor on several occasions as a comparison instrument and was still unable to find the large drop concentrations observed by others.

There is some evidence that the gelatin used by the Meszaros was not recording small droplet impressions. Studies described in Part I showed that even impacted droplets are not always recorded and that drops less than  $5\mu$  diameter are least likely to be recorded in an adverse condition. This may explain why few droplets smaller than  $5\mu$  were noted by Meszaros.

Meszaros showed the results of sedimentation and impaction methods as being well correlated down to about  $8.0\mu$  radius. Her impactor data showed a decrease in number of sizes smaller than  $8\mu$ , with a cutoff value at about  $6\mu$ . This cutoff has not been noticed by others who employed gelatin as a medium; however, it was observed in other media which will not record small droplet impressions (9 and 38).

There is poor correlation in the Meszaros (13) data between visibility and liquid water content as formulated by Eldridge (27). He employed

0.35 to  $10\mu$  radius droplets in the concentration range of about 400 to  $8000 \text{ drops cm}^{-3}$ . Eldridge emphasized the importance of small droplets in visibility by showing the increase in transmission with the omission of small sized droplets (less than  $1\mu$  radius).

Meszaros (13) credited the absorption by nuclei of radius equal to or greater than  $0.15\mu$  in the concentration of  $1000 \text{ drops cm}^{-3}$  with the decrease in visibility in fog to 140 m (average of several cases). She calculated visibility (V) to be approximately 2800 m for the corresponding observed distributions. Data (39) indicate that a particulate mass of  $0.002\mu \text{ gm cm}^{-3}$  (liberal estimate from Figure 3 of Meszaros) would allow a greater than 9600 m visibility. Estimating the effect of particulate matter on the total visibility of the Meszaros fog from scattering and absorption one has

$$1/V_{\text{calculated}} = 1/V_{\text{drops}} + 1/V_{\text{particles}}$$

or

$$V_{\text{calculated}} = \frac{1}{1/2800 + 1/9600} = 2160 \text{ m}$$

V calculated in this way is 2160 m versus 140 m observed, or the decrease in V due to dust reduces the calculated visibility (2800 m) by a factor of less than 5 (liberal estimate) when a factor of 20 is needed to correlate observed and calculated V. May (28) viewed the absorption by particulates during fog as playing a minor role in deterioration of visibility in fog because when fog lifts, visibility goes up very markedly even though the same number of nuclei are present in the atmosphere. Thus, there is no alternative but to assume the presence of lower modal values in the distribution of fog droplets or haze from the Meszaros data.

Evans (37) found the mode to be between 5 and  $40\mu$  radius when employing an electrostatic fog collection technique which imparted a charge to droplets and impacted them on a film of PVA. Collection efficiency was given as 65% for the  $1\mu$  diameter droplets; however, reduced counts were obtained when wind exceeded  $1 \text{ m sec}^{-1}$ . The orientation of the instrument was also critical for obtaining drops from cloudy air and therefore the reliability of small droplet concentrations from Evans' data can be questioned.

Although MacCready's values compared well with those of H. Dessens (2), the efficiency of the web technique of Dessens becomes very



poor near  $2\mu$  radius; thus the two evaluations may have been correct to near  $2\mu$  radius, but for droplets smaller than this value either datum is questionable.

Those who employed the Casella impactor with gelatin (11 and 28) found the median of fog drop distributions to lie from below 1 to  $2.4\mu$  radius. Reasons discussed above indicate that the absence of these low values in the Meszaros (13), Horra (36), Evans (37), and MacCready (2) data was due to media or instrumentation.

#### Web, Transmission Methods

The validity of the spider web method in collecting droplets down to  $2.5\mu$  radius was borne out by simultaneous visibility and drop size distribution measurements which were in close agreement (25, 26).

Eldridge (31) calculated that the number of drops that must have escaped capture by Arnulf's spider web technique was greater than 1000. This estimate was based on computer-determined distributions (40) and Junge's haze distributions (41). Eldridge (27) also employed infrared transmission techniques, correlating drop size with transmission measurements at several wavelengths.

#### All Media and Methods

The number of droplets estimated by Eldridge (27, 31) seems rather large (thousands  $\text{cm}^{-3}$ ) compared to other values given in Table I. Dessens (25) correlated his measured distributions with visibility measurements and found only about 100 droplets  $\text{cm}^{-3}$ . Dessens reasoned that drops of small radii (less than  $2.5\mu$ ) which were not caught by his spider web would not contribute significantly to reduced visibility. Subsequently, it was shown that this hypothesis was incorrect, using transmission calculations based on distributions of Eldridge (27). In fogs that he investigated, May (28) indicated that the existence of more small droplets was doubtful. He showed that the attenuation due to the small droplets he collected rarely accounted for more than 10% of the total attenuation and that in general an error of 10 times their number would be necessary for matching the observed and calculated visibilities.

Examination of fog near the coast almost invariably revealed some modes between 7 to  $10\mu$  radius (34, 35, 38). Therefore, it is possible that the primary mode was not observed because many droplets were below detection limits of methods employed. It is also possible that sea fog actually exhibits a mode between 7 to  $10\mu$  radius.

Evidence that the primary mode may have been missed is abundant; but there are no data showing a fog drop distribution mode from 7 to 10 $\mu$  that correlate these distributions well with visibility.

Eldridge (27) indicated the possible lack of detection of small droplets in some fog drop size distribution studies. When oil was used, Kojima's (33) observation of a mode shift with evaporation of small drops into the oil or atmosphere can account for some of the observed modes near 7 to 10 $\mu$ . The data of Keily (23), Diem (20), Eldridge (27), Arnulf (26), MacCready (2), Houghton (35), Homma (36), and Meszaros (13) showed a mode in the 7 to 10 $\mu$  radius interval. The first four authors showed an approached mode below 2 $\mu$  while the others noted only the mode at 7 to 10 $\mu$ . Reasons given in the text indicate that collection methods or capture media could be at fault in the remaining works, causing a great number of small droplets to be overlooked.

#### B. Conclusions

Based on data reviewed here, visual measurement allowed size determinations to 0.5 $\mu$  radius with 1300 droplets  $\text{cm}^{-3}$  maximum (28) while indirect methods showed sizes to 0.35 $\mu$  diameter with approximately 10,000 droplets  $\text{cm}^{-3}$  maximum (31). Only one published article (28) listed thousands of observed droplets  $\text{cm}^{-3}$  in the range 0.45 to 0.75 $\mu$  radius by visual methods, although Eldridge postulated that greater than this number would be present in all fogs, based on transmission measurements.

It appears then that either a great number of small droplets are missed by collection and replication, and/or factors other than scattering are important sources of transmission attenuation in fog.

### PART III

#### DESCRIPTION OF EXPERIMENTS AND ANALYSES

With knowledge of the limitations of drop size distribution measurement techniques, two methods were selected for use. Thin Formvar and a vacuum impactor with oil were employed in experiments at Ft. Rucker, Alabama, during radiation fog. Formvar, gelatin, and vacuum impactors with fluid were used to document drop size distributions in advection fog at Arcata, California. Gelatin and Formvar slides were taken during an experiment as a comparison between the Formvar and gelatin media.

Experiments on the number and size of drops grown by hygroscopic particles produced by flares allowed determination of the growth and fall-out time (distance) of droplets.

##### A. Measurement of Fog Drop Size Distributions

Samples of the radiation fog at Ft. Rucker, Alabama, were made on 13 May 1968. This fog was sampled using the Formvar solution technique by Mr. J. R. Hicks of the U. S. Army Terrestrial Sciences Center (5). A drop of 5% Formvar in chloroform was spread on a standard microscope slide by spreading with another slide. Slide exposure time was 30 seconds.

Samples of advection fog droplets were taken at Arcata, California in June of 1968, employing the vacuum impactor with silicon fluid (9000 centipoise) and natural settling on gelatin and Formvar-coated standard microscope slides. Use of the vacuum impactor with a 100 cm<sup>3</sup> sample chamber revealed few drops. During the days of fog when vacuum sampling was carried out, only about 10 drops in 100 cm<sup>3</sup> of air were found. These were from 10 to 40μ in diameter.

Early tests with gelatin slides which had been prepared prior to the experiment did not record drop impressions well. Formvar-coated slides were employed for the next few days of fog, and then simultaneous Formvar and fresh gelatin slides were used. Because of the variable drying time of Formvar solutions and the evaporation of droplets due to low relative humidity present at Arcata, gelatin was employed as the collecting medium in the remainder of the tests. The slides taken at Arcata were exposed for one minute.

Weather conditions, sampling techniques and types of tests are summarized in the following table. Visibility (V) is given in meters, wind

direction (dd) in tens of degrees from north, and speed (ff) in meters sec<sup>-1</sup>.

TABLE II. Summary of Data Applicable to Fog Droplet Samples

<u>Location</u>	<u>Date/Time</u>	<u>Slide Type</u>	<u>R.H.(%)</u>	<u>V(M)</u>	<u>dd/ff</u>	<u>Remarks</u>
Ft. Rucker	5/13,0545	Formvar(F)	100	300	00/0.5	Dense clearing fog.
Arcata	6/18,0843-0910	F	94	800	29/2.5	
Arcata	6/24,0810-0900	F	100-86	200 21000	00/00 28/02	Very fast clearing.
Arcata	6/25,1220	Gelatin(G)	92	2500	24/5.5	
Arcata	6/26,0545-0549	G	96	1000	00/02	
Arcata	6/26,0615	G	-	-	-	Start "salty frog"* burn.
Arcata	6/26,0617	G	-	2000	-	Under "frog" smoke.
Arcata	6/26,0628	G	-	-	-	Down runway from "frog" under smoke.
Arcata	6/26,0630	F	-	-	-	In smoke on runway.
Arcata	6/26,0635	G	-	-	-	In smoke on runway.

#### Ft. Rucker Fog Drop Size Distribution

It was hoped that the number ( $n_i$ ) of droplets cm<sup>-3</sup> in a given size interval could be calculated from the following formula:

$$n_i \text{ cm}^{-3} = n_i \text{ counted} / \text{Area counted} \times \text{terminal velocity of drop} \times \text{time slide exposed.}$$

\* See following text for definition.

However, owing to the very low terminal velocities of the small drops counted, the formula gave inordinately high values of small drops.

Another method of making an approximation was employed. Wind speed was approximated at  $0.5 \text{ m sec}^{-1}$  and the small drops were assumed to move with the air. Using an efficiency of 100% for all droplets gave the distribution in column 3 of Table III.

The fog was slowly evaporating and reduced visibility to 300 m. An estimate of the fog drop size distribution from Eldridge's data (31) is indicated in column 4 of Table III.

TABLE III. Fog Drop Parameters of a Radiation Fog at Ft. Rucker, Alabama

$r(\mu)$	$K_s$	$n \text{ cm}^{-3}$ (observed)	$n \text{ cm}^{-3}$ (Eldridge, evolving fog, Dist. 5)	$r^2 K_s (10^{-8}) n$
0.35				
-0.5	3.8	12	1800	1800*
1	2	44	100	88
1.5	2.8	39	30	254
2	2	37	15	296
2.5	2.5	30	4	180
3	2	20	4	400
3.5	2	12	5	240
4	2	3	15	96
4.5	2	7		308
5	2	8	25	400
5.5	2	0		0
6	2	2		160

Except for small sizes, the size and number are in close agreement in the experimental values and those of Eldridge.

Calculation of visibility is possible from Koschmieder's formula (42),

$$V = 3.95/\sigma$$

\* Estimated, see text.

where  $\sigma$  is  $2\pi r_i^2 K_s n_i$ ,  $r_i$  is the average radius in the interval under consideration,  $n_i$  is number per volume in the interval, and  $K_s$  is the corresponding scattering coefficient. Values of  $K_s$  were approximated from Houghton and Chalker (43) at a wavelength of  $0.55\mu$ . Using the distribution in column 3,  $V = 600$  m. Assuming the low number of  $0.5\mu$  size is due to poor collection efficiency and including the estimate of  $1800 \text{ cm}^{-3}$  from Eldridge's evolving fog distribution 5, where  $V = 0.29$  km, the calculated visibility ( $V_c$ ) is equal to approximately 300 m. The liquid water content (lwc) calculated in the conventional way using the observed distribution

$$\text{lwc} = 4/3\pi \sum_i r_i^3 n_i = 0.02 \text{ gm}^{-3}.$$

The omission of droplet sizes below  $0.5\mu$  radius contributed only 5% of the lwc in the above case; however, this omission caused calculated visibility to be incorrect by 100%.

#### Arcata Fog Drop Size Distribution

Distributions of fog droplets recorded on 18 June and 24 June 1968 at Arcata, California, are shown in Figures 10 and 11. These results are typical of distributions observed at Arcata, California, during morning hours of high relative humidity and near noon when relative humidity was less. The morning fog contained more small droplets with few large while the noon advection fog contained more large droplets. In both cases calculated ( $V_e$ ) and observed ( $V_o$ ) visibilities were different by a factor of 2 or greater. Perhaps this discrepancy can be attributed to poor collection of small drops.

To test the usefulness of pyrotechnically produced hygroscopic nuclei (44), a 240 g flare of NaCl and LiCl in a pyrotechnic mixture was burned. The pyrotechnic flare was referred to as a "salty frog". The heat of the burning "salty frog" flare and the heat of condensation of the condensed water on the hygroscopic components caused the pyrotechnically produced salts to rise. In all cases the nuclei thus produced and their condensate caused a decrease in visibility.

Since laboratory studies have shown the poor recording properties of gelatin-coated slides for droplets below  $5\mu$  radius when sampling by natural settling, the number of the droplets in this range can only be estimated from the measured distribution of other sizes. The drop size distributions during the test are shown in Figure 12.

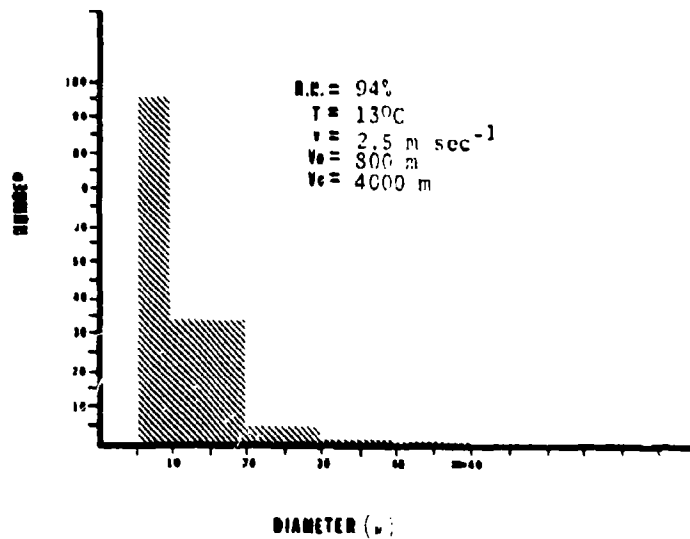


FIGURE 10. DROP SIZE DISTRIBUTION, JUNE 18,  
0343 - 0910 HRS., AT ARCATA.

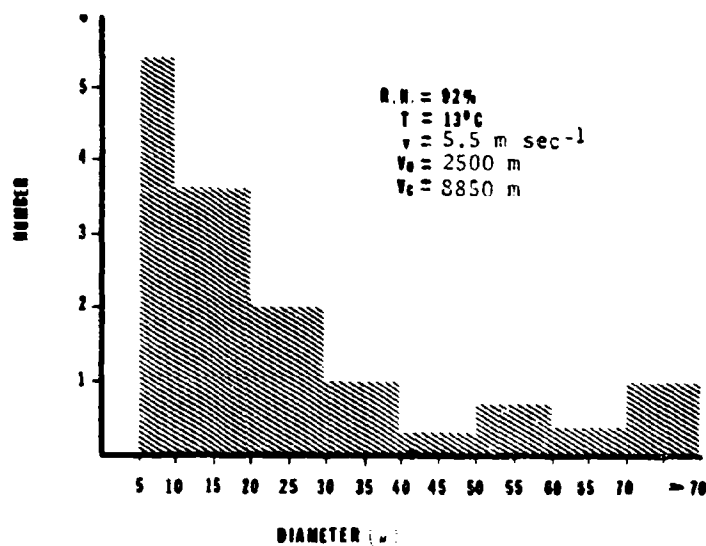
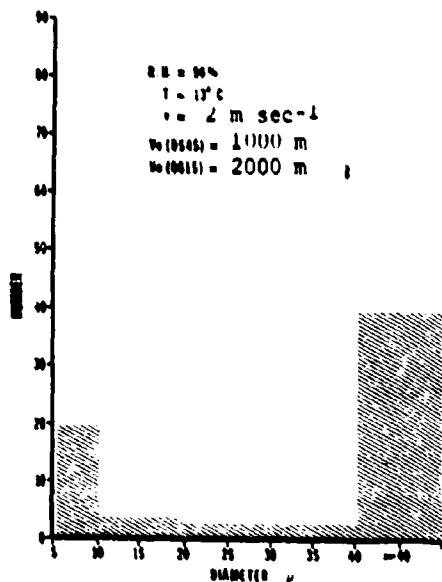
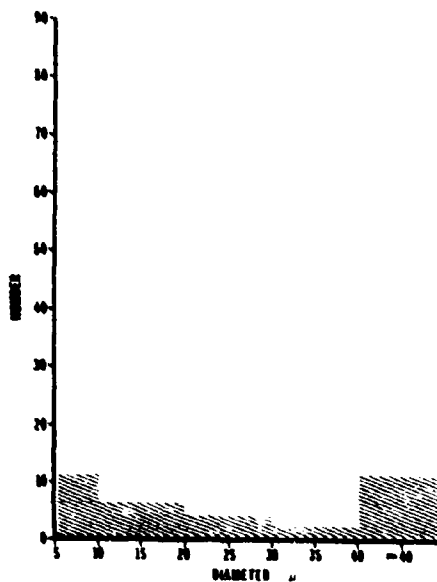


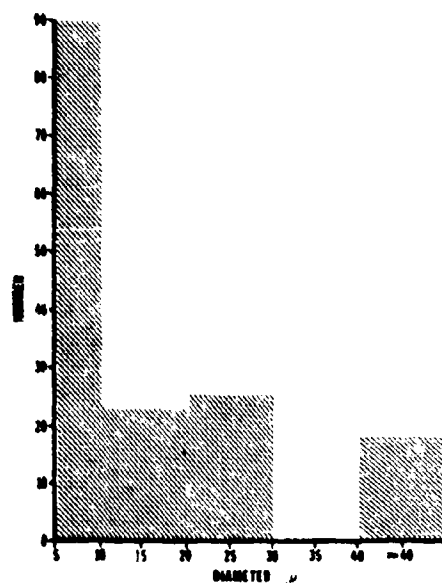
FIGURE 11. DROP SIZE DISTRIBUTION, JUNE 25,  
1220 HRS., AT ARCATA.



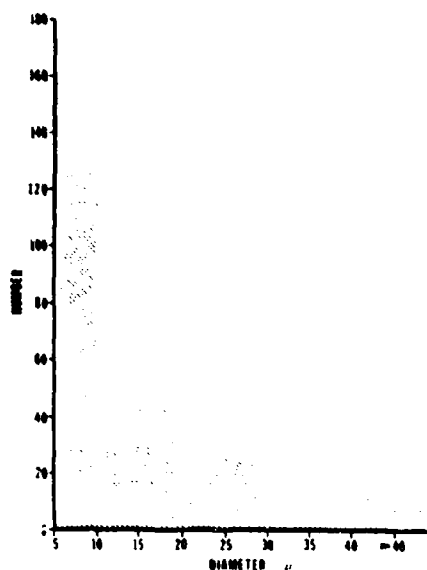
A. 0545



B. 0628



C. 0630



D. 0635

FIGURE 12. CHANGE IN DROP SIZE DISTRIBUTION AFTER SEEDING WITH PYROTECHNIC NUCLEI



The number of droplets in the 5 to 10 $\mu$  diameter range is low in the distributions measured for 0545 to 0628 hours (Figures 12A and 12B). The increase in the small size fraction of droplets in the slides taken at 0630 and 0635 (15 and 20 minutes after ignition of the "salty frog") is evident in Figures 12C and 12D. Based on the size distributions of several pyrotechnically produced particles (particle mode diameter 0.2 $\mu$ ) which grow due to the condensation of water to a size of about 2 to 3 $\mu$ , and Figures 12C and 12D, the estimated number of droplets of diameter below 5 $\mu$  is very large. The numerous droplets of this and smaller sizes contribute greatly to reducing visibility, and their effect was pronounced in the vicinity of burning pyrotechnics.

#### B. Particle Size Distribution Downwind From Pyrotechnic Flares

Although some measurements of the particle size produced by pyrotechnic flares have been made, no studies of the growth and dispersion of these particles have been made under field conditions. Testing the pyrotechnics in nonfoggy but humid air allowed an estimate of particle growth without having to estimate which droplets were natural and which were artificially produced.

On 27 June, four types of pyrotechnic flares were burned on the beach near Arcata to determine their effectiveness in condensing water from the atmosphere and to estimate the diffusion of particles in a field condition. Relative humidity was 89%, wind speed was 1.1 m sec<sup>-1</sup> blowing toward the beach from the sea. The direction and speed of the wind was determined by a portable anemometer and its varying direction over the terrain by a simple method. Soap bubbles were released from a location on the beach and these were carried along by the wind. Thus the turbulence, wind speed and direction could be estimated with distance.

Three participants in the experiment stationed themselves downwind of the pyrotechnic burn site at distances of 2, 5, and 14 m. Each participant held a gelatin coated microscope slide perpendicular to the path of each burning pyrotechnic smoke for one minute.

The four pyrotechnics were as follows:

Designation	Major Components	(Supplied by Manufacturer)	
		Particle size mode diameter ( $\mu$ )	No. of Particles per gram pyrotechnic
Olin Matheson X-1118	NaCl	2	-

Olin Matheson X-1121	$\text{CaCl}_2$	5	-
Crane Ammunition Depot WF-9	$\text{MgCl}_2, \text{NaCl}$	0.2	$10^{14}$
Crane Ammunition Depot WF-10	$\text{AlCl}_3, \text{NaCl}$	0.2	$10^{14}$

Although it was difficult to observe, a light fog rolled in from the sea. Before and after the burn, a one-minute sample of the fog was taken and the average counts subtracted from the distribution measured during the tests.

An area of  $0.13 \text{ cm}^2$  was counted on each slide to determine the drop size distribution for droplets above  $6\mu$  diameter. A smaller area was counted for the 1 to  $5\mu$  sizes and the counts scaled to the  $0.13 \text{ cm}^2$  area. Droplets were counted only for sizes  $5\mu$  diameter and greater. Both particles and droplets in the 1 to  $5\mu$  diameter range were counted since it was not possible to distinguish between the two in many cases.

The drop and particulate size distributions at the three stations are given for the four pyrotechnics in Figures 13 through 15. Figure 13 shows drop size summed over all three stations versus number of particles or drops downwind of 4 pyrotechnics. WF-9 produced more of almost all sizes, the exception being in the 1 to  $5\mu$  diameter size which was greater during the burn of the WF-10 pyrotechnic. The WF-10 produced more small (1 to  $5\mu$  diameter) particles and drops and fewer large (greater than  $10\mu$  diameter) drops than any of the other pyrotechnics. The NaCl and  $\text{CaCl}_2$  flares were similar to each other; however, there were more large (greater than  $30\mu$  diameter) drops during the burn of the  $\text{CaCl}_2$  flare than during the NaCl flare.

In Figure 14, distance from the pyrotechnic versus total number of particles and drops is shown. During the burn periods at 14 m more WF-9 particles or drops were found than in the case of the other three pyrotechnics. The  $\text{CaCl}_2$  flare produced fewer particles or drops at any distance.

Figures 15A through 15D show the variation in particle and drop size with distance from the source of nuclei.

The NaCl size distribution change is more desirable than those obtained with the other pyrotechnics. That is, the number of small particles decreased while the number of large drops increased with

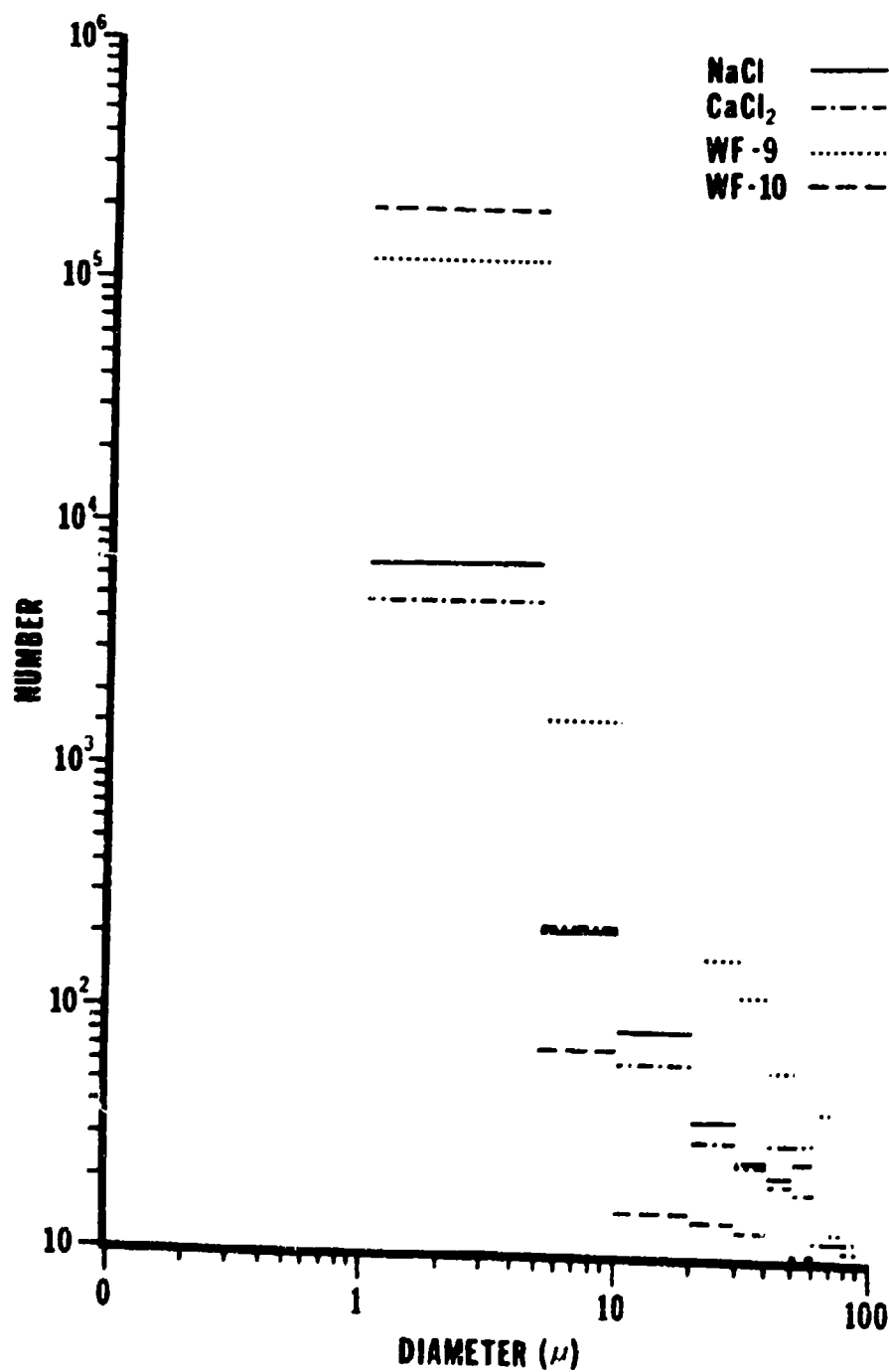


FIGURE 13. NUMBER OF DROPS OR PARTICLES VERSUS DIAMETER

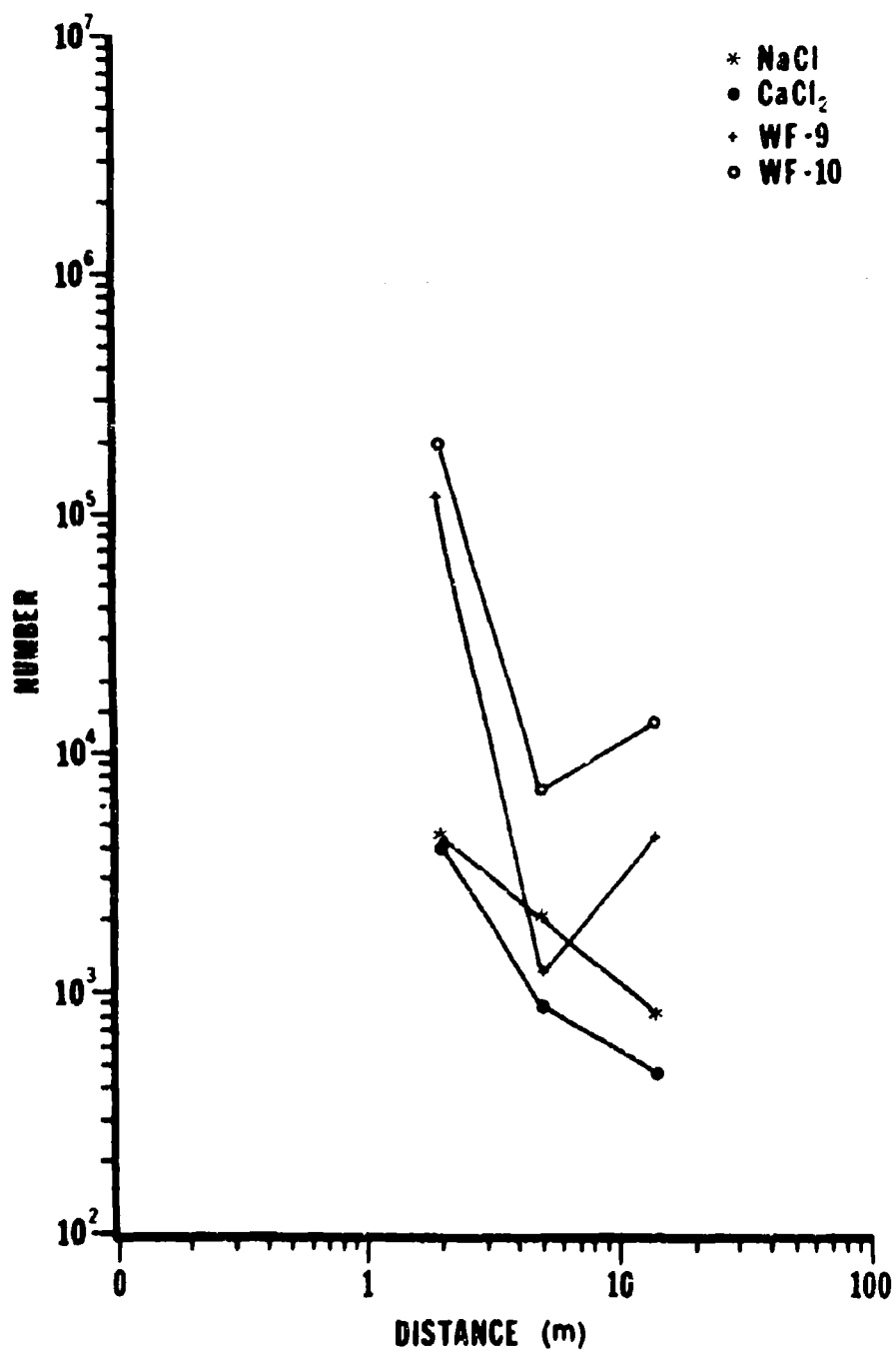
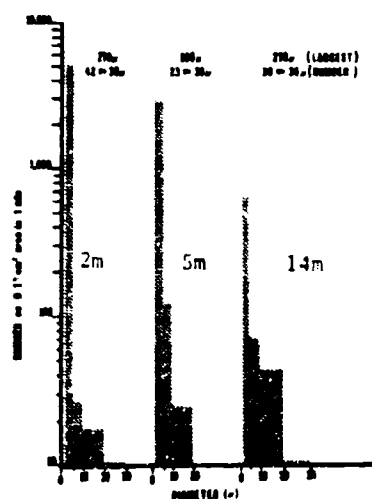
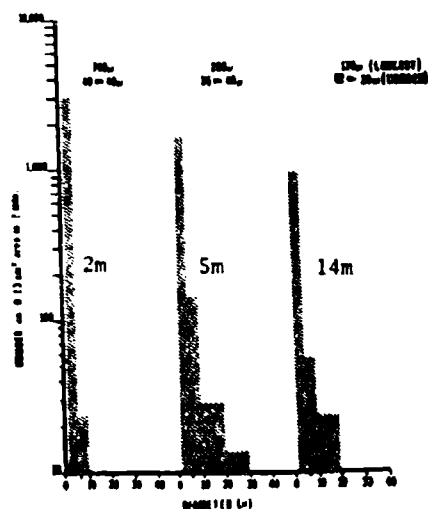


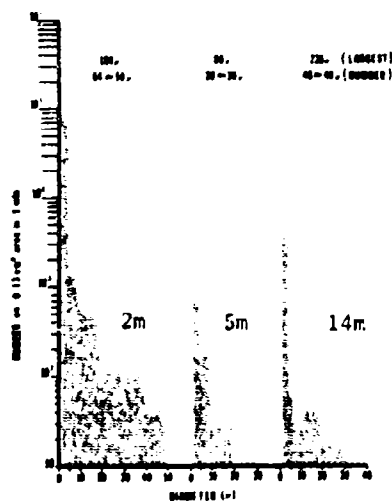
FIGURE 14. NUMBER OF DROPS OR PARTICLES VERSUS DISTANCE



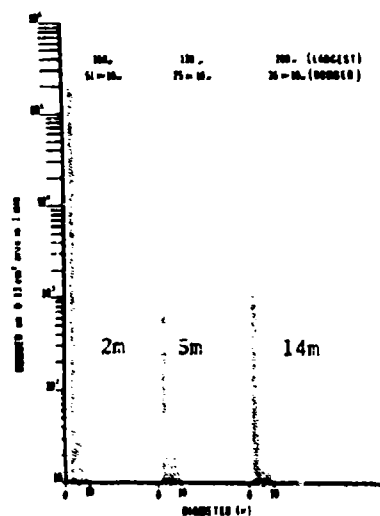
A. NaCl (X-1118)



B. CaCl<sub>2</sub> (X-1121)



C. MgCl<sub>2</sub>, NaCl (WF-9)



D. AlCl<sub>3</sub>, NaCl (WF-10)

FIGURE 15. SIZE DISTRIBUTION VERSUS DISTANCE FROM PYROTECHNIC FLARES

distance from (time after) burn, and the large drops ultimately fell to the ground. This behavior is not discernible in the WF-9 and WF-10 data (Figures 15C and 15D). It could be that the large sizes in the 2 m sample had fallen out by the time the smoke reached the 5 m station and that the particles or drops in the 1 to 5  $\mu$  range had grown into larger sizes by the time the smoke reached 14 m.

The  $\text{CaCl}_2$  pyrotechnic produced the least smoke while the WF-9 pyrotechnic produced the most smoke and eye irritation.

### C. Conclusions

Correlation of drop size distribution with visibility when employing natural settling method with Formvar as a recording media was fair. Assumption of the presence of large numbers of small droplets as postulated by Eldridge yielded good agreement between calculated and actual visibility. Use of gelatin media to record fog droplet impressions always led to higher calculated visibilities than observed.

The use of pyrotechnically created hygroscopic particles as fog dispersing materials resulted in the deterioration of visibility and the increase of small sized droplets.

The WF-9 flare was most effective in producing large drops in the distance examined; however, the presence of the large number of small particles would prevent any increase in visibility which may have resulted from the removal of water droplets from an actual fog. The WF-10 flare was the poorest from both aspects: it produced too many small particles and caused very few large drops.  $\text{NaCl}$  and  $\text{CaCl}_2$  fell between these two extremes. The  $\text{CaCl}_2$  pyrotechnic produced the least number of small particles, caused the least impairment to visibility, and after WF-9 had the highest number of large drops. For these reasons, the  $\text{CaCl}_2$  pyrotechnic was considered the best of the four pyrotechnics tested for fog dispersal purposes.

## SUMMARY

A literature survey revealed that fog drop size distributions were method dependent. A laboratory study showed that neither impaction nor natural settling was entirely reliable. Capture or replication media which included PVA, gelatin, oil and silicon fluid do not always record droplet impressions or retain small droplets for lengthy periods. Lack of correlation between number of captured droplets and visibility was present in even the most careful determinations employing impaction methods. It appears that either small droplets have not been captured and replicated or that factors other than scattering by water droplets contribute heavily to the deterioration of visibility in fog.

Pyrotechnically produced hygroscopic particles caused a persistent decrease in visibility in fog accompanied by an increase in the small droplet sizes fraction. The large number of small particles or droplets produced by four pyrotechnics, tested during a nonfog condition, would negate any visibility improvement brought about by removal of large water drops.

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13. ABSTRACT  In connection with evaluating fog modification efforts, several published methods of measuring the size distribution of fog droplets were reviewed. An evaluation cast doubt on the validity of the gelatin, Formvar, and polyvinyl alcohol, and oil collection media for recording droplets less than 4 $\mu$ diameter. Efficiency of several collection methods appeared to be poor for less than 4 $\mu$ diameter droplets when lack of drop size distribution correlation with visibility measurement was considered.  During a fog dispersal test, pyrotechnically produced hygroscopic reagents were observed to cause an increase in the number of concentrations of the small sized droplets of a natural fog.  During nonfog conditions, several types of pyrotechnic flares containing different hygroscopic reagents were tested for usefulness as fog modification agents. Measurement of drop and particle size distributions downwind from the ignition point of these pyrotechnics revealed that the effect of small droplet sizes in decreasing visibility in air of high humidity would negate the improvement made by removal of large size drops.			

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